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A comprehensive study, culminating in a Reviews of Modern Physics article, was made of semiconductor microcavities exhibiting vacuum-field Rabi splitting. I.e., nonperturbative coupling between the quantum-well exciton absorption resonance and the single optical mode of the cavity. A large number of samples were grown by MBE with various mirror reflectivities and number of quantum wells. Exhaustive measurements were made of linear and nonlinear transmission and reflectively, and photoluminescence using cw and femtosecond lasers as a function of detuning. Samples were grown with record splitting-to-linewidth ratios. A new nonlinear behavior, namely loss of transmission with little change in splitting, was demonstrated and found to arise from excitation-induced dephasing that occurs at lower carrier densities than loss of oscillator strength. A dramatic crossover of the upper and lower branch photoluminescence intensities was shown to occur at the transition from nonperturbative to weak coupling, disproving the claim that it was an exciton polariton laser or boser. Reflectivity linewidths of the coupled system were found to be quantitatively understandable by measuring the quantum-well susceptibility and using it in a transfer matrix computation of the propagation through the cavity, disproving claims that disorder within the quantum wells has to be treated on an equal footing with propagation.

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# FINAL REPORT

# **Controlling Spontaneous Emission of Semiconductor Microcavities**

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## **Executive Summary**

#### **Summary of the Most Important Results**

Nonlinear Behavior of Normal Mode Coupling in Semiconductor Microcavities. We have grown InGaAs/GaAs quantum wells with very narrow linewidths (1 meV=0.6 nm). By growing a single QW in the center of a one-wavelength GaAs spacer or two in the two central anti-nodes of a  $3\lambda/2$  microcavity, we have observed record splitting-to-linewidth ratios (>10, showing that the narrow QW linewidths are also achieved within the microcavity spacer) of so called vacuum Rabi splitting at 4K. This phenomenon is also known as normal mode coupling (NMC) because it exhibits the characteristic anti-crossing curve of two coupled oscillators (here exciton and cavity) as their relative detuning is scanned through zero. We have increased the splitting, seen in reflectivity or transmission, by placing the microcavity in a magnetic field up to 12T, showing that the splitting depends linearly on the electric dipole moment. We have also seen record depth of modulation vacuum field Rabi oscillations because of the high quality of our samples.

We have studied the nonlinear transmission and reflection of our NMC microcavities both by cw pump/probe [Jahnke et al.96] and by femtosecond reflection and upconversion [Lyngnes et al. 97]. Because of our narrow linewidths, we see a new nonlinear behavior: the transmission drops to zero with almost no reduction in NMC; this carrier is explained by nonlinear dispersion theory using the measured exciton absorption with increased density in a transfer matrix formalism. The basic physics is exciton broadening before loss of oscillator strength; as the exciton broadens the absorption increases at the detuned wavelengths of the NMC transmission peaks thus decreasing the Fabry-Pérot transmission (a little bit of absorption in a high-finesse cavity destroy its transmission). Since the area under the exciton absorption (proportional to oscillator strength) changes very little as the broadening sets in, there is almost no reduction in splitting while the transmission drops to zero [Gibbs et al. 97]. This curious nonlinear behavior could not be seen by other groups whose NMC samples were so inhomogeneously broadened that exciton broadening had little effect and loss of oscillator strength dominated the nonlinear behavior. The cw nonlinear measurements and agreement with a many-body theory (with Stephan Koch, Frank Jahnke, and Mackillo Kira, Marburg, Germany) have been published in Physical Review Letters [Jahnke et al. 96] and Optics and Photonic News [Jahnke et al. 97].

The measurements above were performed at 4K, but recently we have achieved record room-temperature normal-mode coupling using

InGaAs/GaAs quantum wells in a microcavity with aluminum-oxide/GaAs mirrors. Normal-mode coupling can be seen at moderate carrier densities even at room temperature [Nelson et al. 96].

Record Normal-Mode-Coupling Microcavities Are Still Far from the Quantum Statistical Limit. The appearance of new frequencies with increased excitation of an NMC microcavity was attributed to climbing a quantum ladder. We concluded that this could not be true by determining that the number of excitation photons that have to be absorbed to see changes in the transmission of a normal-mode-coupling microcavity is about 10<sup>5</sup>. Thus, the best of present-day microcavities are far from the quantum statistical limit. Just as it took atomic physicists more than a decade to reach that limit after first seeing normal-mode coupling, much work remains before semiconductor physicists will achieve quantum entanglement. [Wick et al. 96; Jahnke et al. 96; Gibbs et al. 97; Khitrova et al. 98 & 99]

Crossover of the Upper and Lower Polariton Photoluminescence Intensities is Not Boser Action. When the single mode of a normal-mode-coupling microcavity is tuned slightly higher in energy than the exciton resonance of the quantum wells in its spacer, its photoluminescence following pulsed above-stopband excitation exhibits a curious effect. As the excitation energy increases, the upper polariton luminescence increases much faster than the lower polariton's; as a result, whereas at weak excitation the upper polariton's intensity is much weaker, it becomes much stronger. This threshold like behavior and crossover of the intensities were attributed to boser action, i.e., emission following phonon-assisted Bose-Einstein light condensation into the upper branch. We conducted several definitive experiments. Comparison of cw transmission with the luminescence from both branches as a function of excitation level showed that the transmission is already single-peaked at crossover. Measurement of the normal-modecoupling splitting versus excitation level revealed that the splitting has already collapsed to the detuning by crossover. Determination of the excitation level for lasing established that the lasing threshold density is only a factor of two above the crossover density. From these three experiments, we conclude that the crossover density is too high for boser action to be occurring. Many-body fully quantized calculations show that the observations can be explained as electrons and holes recombining in a microcavity whose emission properties are affected strongly by the exciton absorption within its spacer. These measurements and computations do not rule out the possibility of boser action under more favorable conditions, but they show that the previous claims are wrong. [Kira et al. 97 & 98; Khitrova et al. 98 & 99; Gibbs et al. 97]

Does Structural Disorder Act Differently in Ordinary MQW's than in Bragg MOW's and NMC Structures, Which have in Addition a Strong Light-Matter Interaction? As an experimental test we grew by molecular beam epitaxy a series of NMC structures, and a MQW with QW's identical to those embedded in the microcavities. We extracted the disorder-averaged susceptibility of the QW from a transmission measurement of the MOW sample far detuned from Bragg resonance. By putting the measured QW susceptibility in a transfer-matrix approach for the NMC structures we calculated the reflectivity spectra, and determined the FWHM linewidth as a function of the exciton-cavity detuning. This linear dispersion theory (LDT) procedure assumed one can treat disorder effects separately from the light coupling effects. The comparison to measured reflectivity spectra of the MQW in Bragg resonance and of several microcavities with different cavity finesses and/or different numbers of embedded QW's shows excellent agreement. The good agreement between measurement and LDT shows that LDT works very well for currently studied samples. There are no new properties of the composite system unexpected from the properties of the QW and the empty microcavity put together in LDT, in contrast to the claims that disorder within the QW's has to be treated on an equal footing with propagation. The analysis showed that the linewidths of the two NMC branches are very sensitive to small local changes in the excitonic absorption coefficient and refractive indices in the vicinity of the NMC peaks. The disorder induced asymmetric absorption tails and corresponding asymmetric refractive index changes of the 1s exciton govern the behavior of the reflection dips. Dip equality occurs at a detuning  $E_c > E_x$ , where the excitonic absorption tails are equal at the dip positions. Dip linewidths  $\delta_{upper}$  and  $\delta_{lower}$ are determined mostly by the locally different slopes of the refractive index, resulting in  $\delta_{upper} > \delta_{lower}$ . [Ell et al. 98] In general there is no question that the correct approach is to treat disorder, Coulomb interaction effects, and light propagation on an equal footing, i.e., simultaneously. However, this is a very difficult problem whose solution is not required to predict the properties of currently available composite structures of importance in optoelectronic devices.

Can the Spontaneous Emission Lifetime of Carriers be Deduced by Measuring the Cavity-Mode Photoluminescence. A semiconductor microcavity can be operated in both the nonperturbative regime and perturbative regime depending on the excitation conditions. The perturbative regime is governed by an electron-hole plasma. Its spontaneous emission can be enhanced due to the modified photon density of states by the surrounding cavity. We studied the electron-hole recombination following a nonresonant excitation for carrier densities below the lasing threshold. Previous reports found for those conditions a remarkable enhancement of the carrier decay rate by assuming that the measured cavity-mode photoluminescence (PL)

rate is the same as the carrier decay rate. However, in a high reflectivity cavity as studied previously and here, reabsorption of the cavity-mode PL as well as the large guided-mode density of states cause the dominant decay channel to be the guided modes. Since the cavity does not enhance radiative recombination into guided modes, there is little enhancement of the carrier decay rate. Also, reabsorption within the emitting GaAs layer effectively closes a shutter on the cavity-mode PL, making its decay much faster than the carrier decay. This is because the reabsorption increases as the carriers decay, drastically reducing the fraction of the cavity-mode PL that escapes. Thus, measuring PL dynamics in the cavity mode, one actually detects the PL intensity transmitted through a filter with a strong density- and thus timedependent transmission. We avoided reabsorption in our measurements by collecting the PL through the top mirror without multiple reflections using a ZnSe prism, and compared with measurements collecting the PL through the bottom mirror, thus including reabsorption. Reducing the bottom mirror reflectivity from 99.9% to 78.7% we found an increase of the PL decay time of 25% by avoiding reabsorption, while the reabsorption distorted measurements lead to an increase of a factor of 2.5. Computations using semiconductor luminescence equations agree very well with the experimental findings. They also show only a 25% enhancement of the carrier decay rate and confirm that a relatively large fraction of the spontaneously emitted light is channeled into guided modes and therefore not available for emission in the normal direction. Thus, the large enhancement of the cavity-mode PL decay rate with increased mirror quarterwave pairs is not a cavity-QED effect but a shuttering action of the nonlinear cavity as the absorption recovers [Park et al. 99].

Quantum-Well Magnetoexcitons Do Not Exhibit a Phonon Bottleneck That Might Inhibit Quantum-Dot Lasing. Because the energy levels of quantum dots are discrete, it was suggested that electrons and holes might not be able to relax fast enough to provide gain for lasing. We tested for this so-called phonon bottleneck using pulsed excitation of quantum wells in a magnetic field; quantum-well confinement in the growth direction and magnetic field quantization in the plane result in discrete energy levels much like those of a quantum dot formed by 3D quantum confinement. We excited one of our microcavities 150 meV above the exciton resonance (five times the optical phonon energy) and measured the decay in lasing as a function of magnetic field. The dynamics depends sensitively on excitation density and excitoncavity detuning, but shows no magnetic field dependence. Even at magnetic fields as high as 8 T, where the magnetoabsorption spectra exhibit deep and well-separated Landau levels, there is no increase in the time to maximum emission. These results hold over a wide range of carrier densities, from threshold to several orders of magnitude above threshold. The results evidence a fast relaxation which is uninhibited by the quasi-threedimensional quantum confinement, indicating no phonon-bottleneck reduction of the carrier cooling rate. [Berger et al. 97]

Coherent Control of Microcavity-Laser Stimulated Emission. A curious aspect of the dynamics of our normal-mode-coupling samples relates to the spin precession of quantum-well electrons. Operating just above lasing threshold, we observe oscillations of the stimulated emission for circularly polarized excitation with an in-plane magnetic field B. The oscillations originate from gain modulation via the precession of the electron spin about the B axis. The oscillation frequency is twice the Larmor angular frequency  $\omega_L = 2\pi g_e \mu_B B/h$ , where  $g_e$  is the electron Landé g factor. The oscillation period of 40 ps corresponds to g = 0.45; the output oscillates between  $\sigma^+$  and  $\sigma^-$  lasing This oscillation is a form of coherent control where the stimulated emission is synchronized to the spin precession of the electron. [Hallstein et al. 97]

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### Participating Personnel and Advanced Degrees

Galina Khitrova, PI, was promoted to Associate Professor during this grant period. Other people involved with this research are Hyatt Gibbs, Professor, Claudia Ell, Assistant Research Professor, and Martin Hübner, Postdoctoral Research Associate. Graduate students working for this grant are John Prineas, Sahnggi Park, Eun Seong Lee and Peter Brick. Students receiving Ph.D. with partial support by this grant are Ove Lyngnes and Jill Berger as well as Palace Knights Tom Nelson, Eric Lindmark, and David Wick.

#### **Inventions**

None